BNL-74761-2005-JA

BROWNIAN DRIFT-DIFFUSION MODEL FOR EVOLUTION OF DROPLET SIZE DISTRIBUTIONS IN TURBULENT CLOUDS

Robert McGraw and Yangang Liu
Atmospheric Sciences Division
Environmental Sciences Department
Brookhaven National Laboratory
Upton, NY 11973-5000

May 2005

Submitted to
Geophysical Research Letters

Abstract

Effects from turbulence-induced fluctuations in water vapor saturation on the growth of cloud droplets are examined using a Brownian-like diffusion model to represent the condensation growth/evaporation of droplets along the coordinate of droplet size. The model predicts a diffusive broadening of the droplet size distribution with time, tempered by enhanced evaporation-induced drift of droplets to smaller size from vapor depletion, and approach to a stationary size distribution determined by the balance between size-space diffusion and drift. This balance between diffusion and drift is shown to yield simple analytic expressions for the size distribution that are in good agreement with observed size distributions. Monte Carlo simulations of the approach to the stationary limit and of the distribution itself are presented. A key turbulence parameter required by the kinetic potential theory of drizzle formation [McGraw and Liu, 2003; 2004] is estimated using the new results.

1. Introduction

Uncertainties associated with the physical processes governing clouds and precipitation limit both regional weather forecast accuracy and the ability to predict future global climate using computer models [Houghton et al., 2001]. A large component of this uncertainty derives from complications associated with the coupling between cloud turbulence and microphysical processes over a wide range of spatial/temporal scales and droplet size [Shaw, 2003]. Much effort is currently aimed at reducing uncertainty through the development of more robust parameterizations for clouds and precipitation that are microphysically based yet computationally simple enough so as to be suitable for use in regional to global scale models (Rotstayn, 1999; Rotstayn and Liu, 2005). Especially crucial to understanding many cloud-related phenomena such as precipitation, optical properties, and assessment of the climate impact of anthropogenic aerosols through indirect effects related to the tendency for aerosols to alter cloud properties, is knowledge of the cloud droplet size distribution. Recent progress in parameterizations for clouds and precipitation in atmospheric models [Liu and Daum, 2000; 2004; Liu et al., 2004; 2005], indirect aerosol effects [Liu and Daum, 2002; Rotstayn and Liu, 2003; Peng and Lohmann 2003], and rain initiation theory [McGraw and Liu, 2003; 2004] reinforces the need for better understanding of the spectral shape of the droplet size distribution.

Although significant progress in our understanding of the cloud droplet size distribution has been made and a number of models (e.g., stochastic condensation, entrainment and mixing, and systems theory) have been proposed over the last few decades [Baker et al., 1980; Cooper, 1989; Srivastava, 1989; Khvorostyanov and Curry, 1999; Liu et al., 2002; Shaw, 2003], the details of the processes involved are still poorly understood and highly controversial. The long-standing issue of the spectral broadening (observed droplet size distributions are generally much broader than those predicted by the classical uniform model) remains elusive. Furthermore, few studies/models have yielded analytical forms for the droplet size distribution that agree well with observations, providing the need for development of simple microphysics parameterizations [Liu and Daum 2004].

Previous models of stochastic condensation have usually been of the mean field type. In these models a collection of droplets, estimated on the basis of Kolmogorov scaling to be several meters in extent [Shaw, 2003], is uniformly subject to a low-frequency fluctuating saturation tied to the vertical updraft velocity. However, it has been shown that this uniformity places a severe restriction on the degree to which turbulent fluctuations can lead to broadening of the size distribution [Pruppacher and Klett, 1997]. After pointing out that the early stochastic condensation models generally yield droplet size distributions of the Gaussian type while observations tend to follow positively skewed distributions, Khvorostyanov and Curry [1999] derived a more general mean-field equation that yields gamma droplet size distributions under certain assumptions in the low-frequency regime. Nevertheless, it is clear that the low-frequency limit is often not satisfied in clouds, where significant turbulence fluctuations can occur on smaller spatial scales [Shaw, 2003].

An alternative, simulation-based, approach was described by Kulmala and co-workers [Kulmala et al.,1997; Tisler et al., 2005]. This approach captures fluctuations on the smaller spatial scales by sampling the condensation/evaporation trajectories of individual droplets each allowed to experience a different fluctuation history – thus providing a statistical sampling of the droplet distribution. The droplet growth trajectories are assumed to be driven by turbulence fluctuations in vapor saturation. However, effects from vapor depletion (e.g. on slowing of droplet growth and approach to a stationary size distribution) were not included. Furthermore, such detailed Monte Carlo simulations are too complicated to be used in developing cloud parameterizations. Here we present a new simple model obtained by applying the Langevin equation and Fokker-Planck equation to the study of the cloud droplet size distribution. The model, in which the effect of vapor depletion is accounted for, yields an analytical droplet size distribution of the Weibull form.

2. Turbulent condensation and evaporation: the diffusive growth of cloud droplets

Turbulence causes fluctuations in water vapor saturation and, consequently, in the rates of droplet evaporation and condensation growth. Such fluctuations have been shown to play an essential role in broadening of the cloud droplet size distribution and, it has been proposed, in the production of big cloud droplets responsible for the onset of rapid coalescence growth and initiation of precipitation [Kulmala et al., 1997; Tisler et al., 2005]. The resulting cloud droplet size distributions can be modeled with time either by Monte Carlo simulation methods [Tisler et al., 2005] or analytically, as described below, in terms of a Fokker-Planck equation describing the drift and diffusion of droplets along a coordinate of droplet size.

Cloud droplet growth occurs in the diffusion limited regime for which the growth/evaporation rate takes the form:

$$\frac{dr^2}{dt} = k(T)(S-1) = k(T)\left(\langle S \rangle - 1\right) + k(T)\left(S(t) - \langle S \rangle\right) \tag{1}$$

where r is droplet radius, $k(T)$ is a temperature and pressure dependent rate coefficient (to simplify notation we suppress the weaker pressure dependence) that includes coupled heat and mass transfer during growth/evaporation of the drop [Pruppacher and Klett, 1997]. S is the saturation ratio, defined as the ratio of the vapor pressure of the interstitial cloud air to the equilibrium vapor pressure of the drop. The second equality allows for the possibility that the average parcel saturation ratio $\langle S\rangle$ may be other than unity as a consequence of either adiabatic cooling or vapor depletion. We assume that (i) the fluctuations in *S* can be characterized by a finite variance $\sigma_S^{\,2}$,

$$\left\langle \left(S-\langle S\rangle\right)^2\right\rangle = \sigma_S^{\,2}, \tag{2a}$$

with exponential decay of correlation over timescale γ^{-1},

$$\begin{aligned}&\left\langle \left(S(t)-\langle S\rangle\right)\left(S(t+\Delta)-\langle S\rangle\right)\right\rangle = \langle S(t)S(t+\Delta)\rangle - \langle S\rangle^2 \\ &\langle S(0)S(\Delta)\rangle - \langle S\rangle^2 = \sigma_S^{\,2}\exp(-\gamma\Delta)\end{aligned}. \tag{2b}$$

These assumptions agree with the model of Kulmala and co-workers [Kulmala et al., 1997; Tisler et al., 2005], however it is significant that we will not require that the fluctuations in *S* be gaussian. Thus the present analysis should apply even in the face of large non-gaussian fluctuations in *S* from intermittency – a well known property of cloud turbulence [Shaw, 2003]. Estimates for σ_S (on the order of 1%) and for the correlation time, γ^{-1} (from several seconds to tens of seconds) have also been provided [Kulmala et al., 1997; Tisler et al., 2005]. Such short correlation times suggest that fluctuations in *S* are strongly damped over the time scale, τ, estimated below, of significant change in the droplet size distribution. Finally it is assumed (c.f. the second equality of Eq. 2b) that (iii) the fluctuations are stationary in the sense that their statistical properties depend only on the time difference, Δ.

The preceding suggests an analogy to Brownian particle motion with $z \equiv r^2$ playing the role of the spatial coordinate and Eqs. 1 and 2 giving the instantaneous velocity, $v = dz/dt$. The latter has two components: the random fluctuation term in Eq. 1, which contains $S(t)$ and gives rise to diffusion, and the uniform drift term proportional to $\langle S\rangle - 1$. Thus the full problem, including both diffusion and drift, is analogous to the well-studied model of Brownian motion in a field of force. The formal analogy to Brownian motion allows one to write down many key results immediately rather than having to repeat in detail derivations available in standard texts [e. g., Serra et al., 1986; Gardiner, 1985]. We first consider the random growth component, which in the strongly damped regime ($\gamma^{-1} << \tau$) causes droplets to diffuse along the z coordinate with diffusion coefficient given by the product of the variance of the growth velocity fluctuations, $k^2(T)\sigma_S{}^2$, and correlation time [Serra et al., 1985]:

$$D_Z = \frac{k^2(T)\sigma_S{}^2}{\gamma}. \tag{3}$$

3. Vapor depletion and the stationary cloud droplet distribution

Diffusion along the coordinate of droplet size is checked by requirements that the droplet radius be positive and total water (liquid plus vapor) be conserved; droplets cannot grow without vapor depletion. Vapor depletion will be represented here in the simplest mean field sense by assigning an average saturation $\langle S \rangle$ for the cloud parcel under consideration determined self consistently by the methods now described.

Under stationary conditions analytic results for both $\langle S \rangle$ and the droplet distribution itself are readily obtained. For $\langle S \rangle \neq 1$ the first term on the right hand side of Eq. 1 gives a deterministic drift in droplet size with velocity:

$$v_{depl} = \left(\frac{dr^2}{dt} \right)_{depl} = k(T)\left(\langle S \rangle - 1 \right). \tag{4}$$

The combination of diffusion and drift is, just as in the case of Brownian motion, described by a Fokker-Planck equation [Serra et al., 1985]. In present notation:

$$\frac{\partial f}{\partial t} = D_Z \frac{\partial^2 f}{\partial z^2} - v_{depl} \frac{\partial f}{\partial z} = \frac{k^2(T)\sigma_S{}^2}{\gamma} \frac{\partial^2 f}{\partial z^2} - v_{depl} \frac{\partial f}{\partial z}. \tag{5}$$

The stationary condition $(\partial f / \partial t = 0)$ is determined from the balance between diffusion, which tends to broaden the distribution, and increase liquid water content, and drift, which must therefore tend to narrow the distribution, and decrease liquid water content, by reducing droplets to smaller size (i.e. v_{depl} must be negative) for a stationary distribution. From Eq. 5 we readily obtain a Boltzmann distribution in z for the stationary solution:

$$f_\infty(z) = N_D \frac{|v_{depl}|}{D_Z} \exp\left(-\frac{|v_{depl}|}{D_Z} z \right), \tag{6}$$

where $|v_{depl}| = -v_{depl}$ is the magnitude of v_{depl} and normalization is to the droplet number concentration N_D. The liquid water fraction (cm^3 cloud liquid water/ cm^3 air) is obtained as the 3/2 moment of $f(z)$:

$$L = \frac{4\pi}{3} \int_0^\infty z^{3/2} f(z) dz\,. \tag{7}$$

Substitution of $f_\infty(z)$ for specified liquid water content yields the stationary value of v_{depl}:

$$v_{depl} = -\pi \left(\frac{N_D}{L} \right)^{2/3} D_Z = -\pi \left(\frac{N_D}{L} \right)^{2/3} \frac{k^2(T)\sigma_S{}^2}{\gamma}. \tag{8}$$

Reflective of vapor depletion, the theory predicts a uniform shift in average saturation to values below unity. From Eqs. 4 and 8:

$$\langle S \rangle_\infty = 1 - \pi \left(\frac{N_D}{L} \right)^{2/3} \frac{k(T)\sigma_S{}^2}{\gamma} \tag{9}$$

where the subscript on the left refers to the stationary condition. More generally, as describe in connection with the simulations below, v_{depl} and $\langle S \rangle$ are functions of time. In the absence of fluctuations ($\sigma_S{}^2 = 0$) water vapor is in equilibrium with the droplets under these conditions at $S = 1$.

Transforming Eq. 6 from z to droplet radius gives the following positively skewed Weibull distribution:

$$f_\infty(r) = 2\pi N_D \left(\frac{N_D}{L} \right)^{2/3} r \exp\left[-\pi \left(\frac{N_D}{L} \right)^{2/3} r^2 \right], \tag{10}$$

which is similar to that derived from the maximum entropy principle and is a good representation of typically observed cloud droplet size distributions [Liu et al., 1995; Costa et al. 2000].

4. Monte Carlo Simulation

For simulation of Eq. 5 we utilize the equivalent Langevin equation:

$$dz = v_{depl}dt + \sigma_Z dX \tag{11}$$

where ${\sigma_Z}^2 = 2D_Z$ and $dX = \phi\sqrt{dt}$. ϕ is a dimensionless random variable drawn from a standardized normal distribution with zero mean and unit variance, $p(\phi) = (2\pi)^{-1/2}\exp(-\phi^2/2)$.

With these definitions, $\langle dX \rangle = 0$ and $\langle dX^2 \rangle = dt$. Equivalence of Eqs. 5 and 11 is demonstrated in standard texts on stochastic processes [e.g. Gardiner, 1990; Wilmott et al.,1998]. The drift-diffusion processes they describe are frequently encountered (e.g. Brownian motion in a field of force) and well suited to simulation using Monte Carlo methods. Monte Carlo simulations were carried out for *N*-drop samples of growth/evaporation trajectories based on Eq. 11. The droplets interact through the vapor depletion effect, but are otherwise independent. At each time step the drift velocity as adjusted to as to preserve liquid water content close to its externally specified value $L(t)$. Generally, e.g. with a parcel undergoing adiabatic cooling, *L* would be a function of time and N_D would also change with the activation to new droplets or droplet loss. To illustrate the new methods we will assume the simplest case of fixed values for *L* and N_D specified by the initial condition.

Figure 1 shows results from simulations of Eq. 11 for multiple samples of 100 drops each. Time is expressed in units of the distribution relaxation time mentioned above, $\tau = {z_0}^2/(2D_Z)$, where $z_0 = (3/4\pi)^{2/3}(L/N_D)^{2/3}$ is the average radius squared of the droplets, and radius in units of $r_0 = \sqrt{z_0}$. Scaled results are independent of *L*, N_D and D_Z. The model time step was set at 0.001τ ($d\tilde{t} = 0.001$) and simulations carried out to $t = 5\tau$ ($\tilde{t} = 5$). Positive values for the size coordinate are insured by applying a reflective boundary condition at the origin. The top panel shows the cumulative radial distribution (normalized to unity) for the Weibull distribution of Eq. 10 and comparison with results from combining four 100-drop Monte Carlo simulations at different times in the stationary limit near $\tilde{t} = 4$ (400 points total). The bottom panel shows evolution of the relative dispersion ε, defined as the square root of the variance of the droplet size distribution divided by its mean. The points show values of $\varepsilon(\tilde{t}_k)$ at each successive time step, $\tilde{t}_k = 0.001k$, over the course of the simulation. The initial dispersion is zero, corresponding to a monodisperse initial size distribution. This is followed by broadening of the distribution with time, due to the turbulence fluctuations in saturation and condensation growth/evaporation rates, preserving L and N_D. Note that the broadening seen here is counter to the usual tendency of condensation growth at fixed (non-fluctuating) saturation to narrow size distributions over time [McGraw, 1997]. Broadening is effectively complete by $\tilde{t} = 1$ with slower approach to the asymptotic value, $\varepsilon_\infty = \sqrt{4/\pi - 1} = 0.5227...$, horizontal line, determined by the moments of the stationary distribution, Eq. 10. Although not shown here, the drift velocity also approaches the analytic result (Eq. 8).

5. Implications for the study of clouds and precipitation

The simplest Brownian drift-diffusion model (constant coefficients in Eq. 11) has been found to yield a Weibull spectrum of cloud droplets in excellent agreement with shapes and relative dispersions of typically observed cloud droplet size distributions. More general distributions can be obtained by allowing for drift/diffusion coefficients that are a function of droplet size. For example, the Kelvin effect, not included here, gives a small additional, and size dependent, contribution to the drift to smaller droplet size. Alternatively, empirical size distributions can be used and Eq. 5 inverted to obtain information on the drift/diffusion rates. Such extensions of the method will be described a future publication.

The present analysis also provides a key turbulence parameter needed in the kinetic potential (KP) theory of drizzle formation [McGraw and Liu, 2003; 2004]. This is the quantity $t_{1\%}$, defined as the time required for diffusion along the growth coordinate to change the cloud droplet size 1% from 10 to 10.1 micron radius. From the preceding this can now be expressed in terms of the diffusion constant: $t_{1\%} = (\Delta z)^2/(2D_Z)$, where $\Delta z = 10.1^2 - 10.0^2 = 2.01\mu m^2$ and the units of D_Z are $\mu m^4 s^{-1}$. We now estimate D_Z from Eq. 3: Using $k(10^\circ C) = 167.8\mu m^2 s^{-1}$, from Eq. 13.28 and the parameters given in Table 13.1 of Pruppacher and Klett [1997], saturation variance $\sigma_S = 0.01$ and correlation time $\gamma^{-1} = 7s$, both from Kumala et al. [1997], yields $D_Z = 20.2\mu m^4 s^{-1}$ and $t_{1\%} = 0.1s$. This is in the range of previous very rough estimates for this parameter and happens to be a condition for which detailed calculations of the drizzle barrier and drizzle rate have been presented [McGraw and Liu, 2003; 2004]. Finally for a typical mean droplet radius $r_0 = 10\mu m$ ($z_0 = 100\mu m^2$) we obtain the estimate $\tau = z_0{}^2/(2D_Z) \approx 4\min$ for the distribution relaxation time, thus justifying the strongly damped condition ($\gamma^{-1} << \tau$) used in derivation of the Fokker-Planck and equivalent Langevin equations (Eqs. 5 and 11, respectively). In conclusion, the present theory provides both a mechanism for shaping the cloud droplet distribution and foundation for the similar drift-diffusion processes that underlie the KP theory of drizzle initiation.

Acknowledgments: The authors thank Drs. Peter H. Daum and Stephen E. Schwartz of BNL for valuable discussions. This research was supported by the Office of Biological and Environmental Research of the US Department of Energy, as part of the Atmospheric Radiation Measurements (ARM) Program under contract DE-AC02-98CH10886.

References:

Baker, M. B., R. G. Corbin, and J. Latham (1980), The influence of entrainment on the evolution of cloud droplet spectra. I: A model of inhomogeneous mixing, *Quart. J. Roy. Meteor. Soc. 106*, 581-598.

Cooper, W. A. (1989), Effects of variable droplet growth histories on droplet size distributions. Part I: Theory, *J. Atmos. Sci. 46*, 1301-1311.

Gardiner, C. W. (1990), *Handbook of Stochastic Methods* (Springer-Verlag, New York).

Houghton, J. T., Y. Ding, D. J. Griggs, M. Noguer, P. van der Linden, X. Dai, K. Maskell, Eds., *Climate Change 2001: The Scientific Basis. Contributions of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*; (Cambridge University Press, Cambridge, 2001).

Khvorostyanov, V. I. and J.A. Curry (1999), Theory of Stochastic Condensation in Clouds. Part I: A General Kinetic Equation. *J. Atmos. Sci, 56*, 3985-3996; Part II: Analytical Solutions of the Gamma-Distribution Type. *J. Atmos. Sci, 56*, 3997-4013.

Kulmala, M., U. Rannik, E. L. Zapadinsky, and C. F. Clement (1997), The effect of saturation fluctuations on droplet growth, *J. Aerosol Sci. 28*, 1395-1409.

Liu, Y. and P. H. Daum (2000), Spectral dispersion of cloud droplet size distributions and the parameterization of cloud droplet effective radius, *Geophys. Res. Letts. 27*, 1903-1906.

Liu, Y. and P. H. Daum (2002), Indirect warming effect from dispersion forcing, *Nature 419*, 580-581.

Liu, Y. and P. H. Daum (2004), Parameterization of the autoconversion process. Part I: Analytical formulation of the Kessler-type parameterizations, *J. Atmos. Sci. 61*, 1539-1548.

Liu, Y., L. You, W. Yang, and F. Liu (1995), On the size distribution of cloud droplets, *Atmos, Res. 35*, 201-216.

Liu, Y., P. H. Daum, S. K. Chai, and F. Liu (2002), Cloud parameterizations, cloud physics, and their connections: An overview. In *Recent Research Developments in Geophysics*, S. G. Pandalai, Ed., pp. 119-142, (Research Signpost, Trivandrum, India)

Liu, Y., P. H. Daum, and R. McGraw (2004), An analytic expression for predicting the critical radius in the autoconversion parameterization, *Geophys. Res. Letts. 31*, L06121, 10.1029/2003GL019117.

Liu, Y., P. H. Daum, and R. McGraw (2005), Size truncation effect, threshold behavior, and a new type of autoconversion parameterization *Geophys. Res. Lett.*, accepted for publication.

McGraw, R. (1997), Description of aerosol dynamics by the quadrature method of moments, *Aerosol Sci. and Tech. 27*, 255-265.

McGraw, R. and Y. Liu (2003), Kinetic potential and barrier crossing: a model for warm cloud drizzle formation, *Phys. Rev. Letts. 90*, 018501-1 thru 018501-4.

McGraw, R. and Y. Liu (2004), Analytic formulation and parameterization of the kinetic potential theory for drizzle formation, *Phys. Rev. E 70*, 031606.

Peng, Y. and U. Lohmann (2003), Sensitivity study of the spectral dispersion of the cloud droplet size distribution on the indirect aerosol effect, *Geophys. Res. Letts. 30*, 14-1-14-4, doi: 10.1029/2003GL017192.

Pruppacher, H. R. and J. D. Klett (1997), *Microphysics of Clouds and Precipitation* (Kluwer Academic).

Rotstayn, L. D. and Y. Liu (2003), Sensitivity of the indirect aerosol effect to an increase of cloud droplet spectral dispersion with droplet number concentration. *J. Climate 16*, 3476-3481.

Rotstayn, L. D. and Y. Liu (2005), A smaller global estimate of the second indirect aerosol effect. *Geophys. Res. Lett. 32*, L05708, doi:10.1029/2004GL021922.

Serra, R., M. Andretta, G. Zanarini, and M. Compiani (1986), *Introduction to the Physics of Complex Systems*, (Pergamon, New York) chapt 1.

Shaw, R. A. (2003), Particle-turbulence interactions in atmospheric clouds, *Annu. Rev. Fluid Mech. 35*, 183-227.

Srivastava, R. C. (1989), Growth of cloud drops by condensation: A criticism of currently accepted theory and a new approach, *J. of the Atmos. Sci. 46*, 869-887.

Tisler, P., E. Zapadinsky, and M. Kulmala (2005), Initiation of rain by turbulence-induced condensational growth of cloud droplets, *Geophys. Res. Lett., 32*, L06806, doi:10.1029/2004GL021969.

Wilmott, P., S. Howison, and J. Dewynne (1998), *The Mathematics of Financial Derivatives* (Cambridge University Press, Cambridge) Chapt. 2.

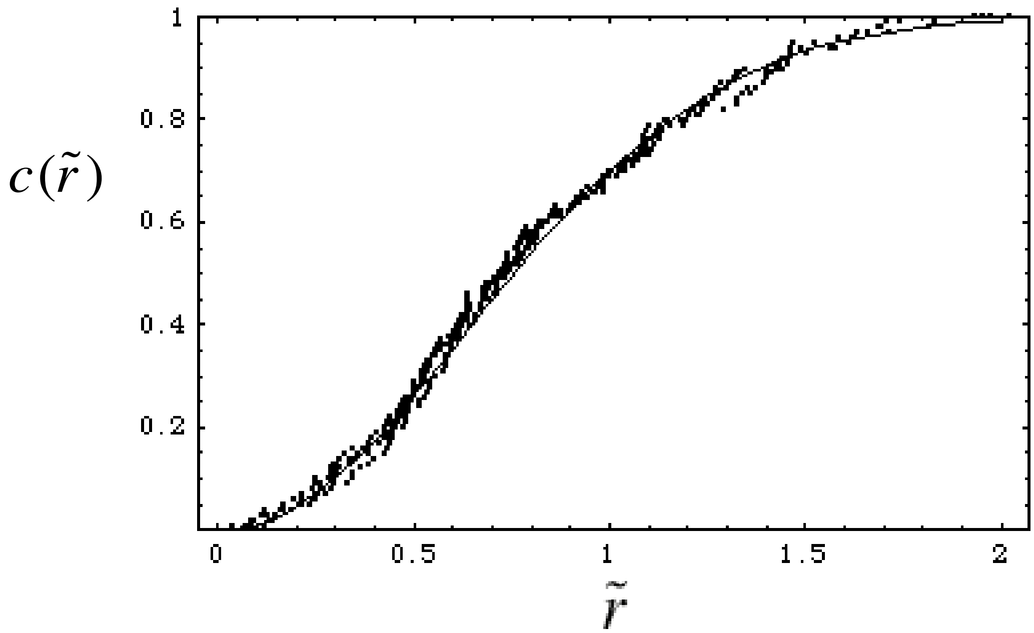

Fig. 1 Top panel

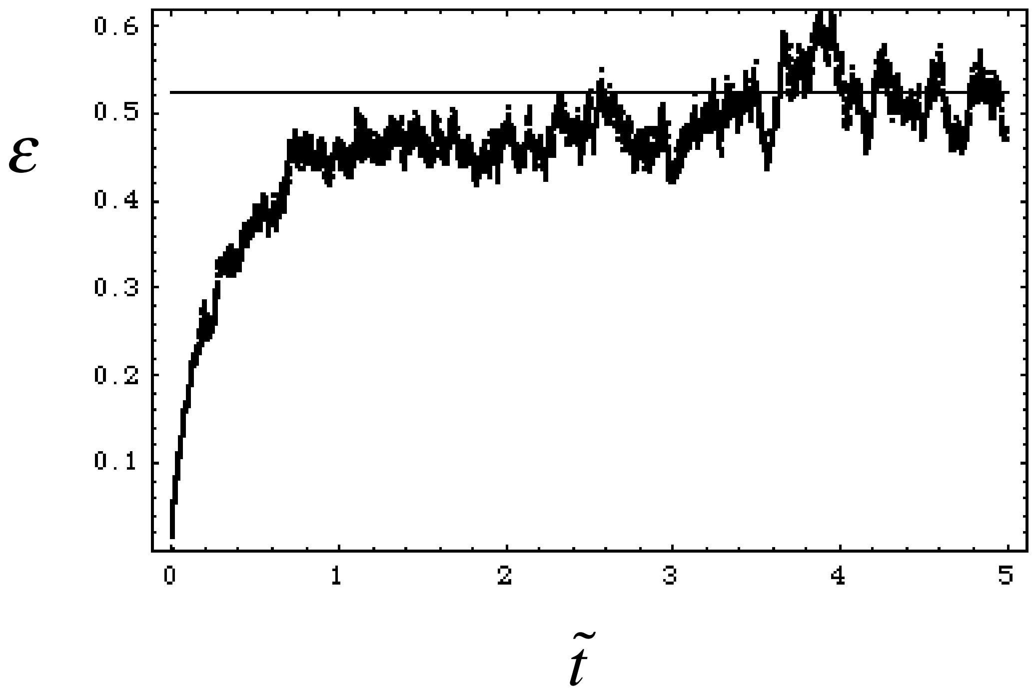

Fig. 1 Bottom panel

Figure 1. (Top) Cumulative radial distribution versus scaled drop radius from Eq. 10 (solid curve) and comparison with results from four 100-drop Monte-Carlo simulations (points) at different times near $\tilde{t} = 4$. (Bottom) Relative dispersion, ε, for a 100-drop sample taken at reduced time increments of 0.001 (5000 samples total) as a function of reduced sample time. Results are shown for evolution from an initially monodisperse size distribution. The fluctuations seen in ε are due to the limited sample size.